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On cavity modification of stimulated Raman scattering

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Abstract
We study theoretically stimulated Raman scattering (SRS) in a nonlinear dielectric microcavity and compare SRS thresholds for the cavity and the bulk material it is made of. We show that cavity SRS enhancement results solely from the intensity build up in the cavity and from the differences of the SRS dynamics in free and confined space. There is no significant modification of the Raman gain due to cavity QED effects. We show that the SRS threshold depends significantly on the nature of the dominating cavity decay as well as on the coupling technique with the cavity used for SRS measurements.

Keywords: Cavity QED, whispering gallery modes, stimulated Raman scattering

1. Introduction

Spontaneous emission processes may be either enhanced or inhibited in a cavity due to a modification of the density of electromagnetic states compared with the density in a free space [1, 2]. The changes in the properties of spontaneous emission play an important role in lasers and lead to 'thresholdless lasing' [3–5]. There is great activity in theoretical and experimental investigations of those and other cavity quantum electrodynamics effects [6–11].

Substantial optical power enhancement within a high-finesse optical cavity has recently yielded continuous wave Raman lasers with low threshold and large tunability [12, 13]. Such properties make cavity-enhanced continuous wave Raman lasers attractive for high resolution spectroscopy, remote sensing, atomic physics, and telecommunications. Reducing the cavity size may further improve the performance of the lasers. Open dielectric spherical microcavities are promising for those purposes.

Various cavity effects, including resonance-enhanced fluorescence [14], were observed with dielectric microsphere resonators [15–17]. A dielectric microsphere possesses natural modes of light oscillations that are usually called whispering gallery modes (WGMs) or morphology-dependent resonances. WGMs may have high quality factors and small volumes which is very important for cavity QED.

An enhancement of stimulated Raman scattering (SRS) is one of the effects demonstrated in spherical microcavities. Low threshold SRS was observed with pulsed [18–20] and continuous wave [21, 22] optical pumping in micrometre-size liquid droplets. Microcavity QED enhancement of Raman gain has been inferred from measurements of a dependence of the SRS threshold on the size and material of the microdroplets, and comparison with the values of SRS threshold reported for liquid core fibers having equivalent interaction length and core composition [21, 22]. This enhancement has been linked to the cavity modification of the properties of usual lasers. A theory of the Raman gain modification that explains the experimental results has been developed [23, 24]. However, recent experiments with silica microspheres have not shown any significant change in SRS gain which might be attributed to quantum effects [25]. It was thought that the quantum effects were not observed due to experimental difficulties with measuring the properties of very small cavities.

The contradiction of the recent and previous experimental results is fundamentally important. Indeed, in a Raman laser both pump and generated radiation may be tuned far away from the corresponding molecular/atomic transitions. Usually, in Raman systems it is possible to eliminate the excited states adiabatically and consider light interaction with degenerate ground states only. A cavity may not usually influence these states. It is not clear, therefore, if the cavity enhancement or inhibition of spontaneous emission may change the SRS process significantly.

Nonlinear optics in microcavities is important not only from the purely theoretical, but also from a practical, point of view. Recently created microcavity Raman lasers [25], for example, represent a route to compact, ultralow threshold sources for numerous wavelength bands that are usually difficult to access. Small WGM cavities made of crystals possessing quadratic nonlinearities result in efficient electro-optic light modulation and microwave photonic reception [26, 27]. Fabrication and handling of cavities with very small size and high quality factors is a complicated technological problem. It is important to find a tradeoff between the importance of the cavity size decrease and the effort spent. Better understanding of size- and Q-factor-dependent cavity enhancement of nonlinear processes is necessary.

We show here theoretically that the increase in the SRS occurs solely due to energy accumulation in the cavity modes. No cavity QED associated Raman gain enhancement exists, unlike the cavity enhancement of the spontaneous emission. We see an explanation of the result in the fact that SRS is essentially a stimulated nonlinear process not influenced by the density of states of the surrounding reservoir. An atom plays the role of a transducer between pump and probe fields with no spontaneous processes involved. The atomic transitions may be off-resonant with respect of the cavity modes. In this case SRS is enhanced due to the classical effect of the field accumulation in the cavity, but no resonant interaction of the atom and the cavity modes occurs, so the cavity modification of the density of quantum states does not change atomic properties.

To study SRS we consider outside-pumped three-level Λ particles embedded either in an optical fiber or in a dielectric cavity made of the same host material. We show that the intracavity SRS is modified in a similar way to a microcavity modification of a laser emission process. This modification, however, does not explain the results of experiments [21, 22]. We propose an explanation of the size dependent efficiency of Raman scattering observed in [21, 22] that attributes the experimental results to a size dependent coupling of a free space light beam to high-Q WGMs. We also show that the size dependence of the SRS threshold varies significantly as the nature of the major cavity losses changes.

2. SRS threshold estimations for a generic active medium

The Raman gain of a Stokes field in an SRS process may be presented as $\exp(g_c I_c L_c)$, where g_c is a cavity Raman gain factor, I_c is an intensity of the pumping light stored in the cavity, and L_c is an effective cavity path length. This expression should be compared with similar expression for the bulk material, $\exp(g_b I_b L_b)$, where g_b is a bulk Raman gain factor, I_b is the intensity of the pumping light in the material, and L_b is a length of the sample. To make the comparison valid we assume that: (i) $L_c = L_b$, (ii) the cavity does not have any absorption losses, and (iii) the input intensity of the pump light for the bulk material and for the cavity is the same.

Let us estimate the threshold pump power necessary for generation of Stokes radiation in an open dielectric spherical cavity. The intensity build up factor for the light resonant with a cavity mode is

$$\frac{I_c}{I_0} = \frac{Q\lambda}{\pi^2 n_0 a}, \tag{1}$$

where I_0 is the intensity of the input light, I_c is the effective intensity of the light in the cavity, λ is the light wavelength, a is the radius of the microsphere, n_0 is the index of refraction of the microsphere host material, and Q is the cavity mode quality factor. We assume that the effective cross-sectional area ($\mathcal{A}$) is the same for the input beam and for the WGM.

The effective interaction length in the cavity is

$$L_c = \frac{Q\lambda}{2\pi n_0}. \tag{2}$$

Assuming now that the pump and the Stokes waves are both resonant with the WGMs of the cavity which have quality factors Q_p and Q_S respectively we derive the condition for Raman amplification (round trip Raman gain exceeds round trip losses)

$$g_c \xi I_0 \frac{Q_p \lambda_p}{\pi^2 n_0 a} > \frac{2\pi n_0}{Q_S \lambda_S}, \tag{3}$$

or, assuming that the mode volume is $V_m \approx 2\pi a \mathcal{A}$ and the pump power is $P_{01} = I_0 \mathcal{A}$,

$$P_{01} > \frac{\pi^2 n_0^2}{\xi g_c Q_S Q_p} \frac{V_m}{\lambda_p \lambda_S}, \tag{4}$$

where $\xi < 1$ is a numeric parameter that describes the noncritical coupling of the pump to the mode as well as the non-perfect overlap between the pump and Stokes modes, and $\mathcal{A}$ is the effective cross-sectional area of the beam. Expression (4) corresponds to the expression presented in [25] if $g_c \equiv g_b$.

Equation (4) is purely classical. It does not take into account an enhancement of Raman gain due to cavity QED effects which might be present in the system if the analogy between enhancement of stimulated emission [3] and SRS works. Two hypotheses of cavity QED enhancement of Raman gain were proposed.

A rough single dimension estimation of the enhancement gives a value equal to the ratio of the free spectral range of the cavity and the width of the Raman gain [21]:

$$\frac{g_c}{g_b} \approx \frac{c}{\pi^2 n_0 a \mathcal{G}} \tag{5}$$

if $c/(\pi^2 n_0 a \mathcal{G}) > 1$, or $g_c = g_b$ if $c/(\pi^2 n_0 a \mathcal{G}) < 1$, where $\mathcal{G}$ is the homogeneous linewidth of the bulk Raman gain. As one might see from (5), resonantly enhanced Raman gain is possible for comparatively small spheres, while the gain is equal to the gain in the bulk for large spheres. Hence, in the case of comparatively small cavities, equation (4) should be modified according to equation (5) as

$$P_{02} > \frac{\pi^4 n_0^3}{\xi Q_S Q_p} \frac{V_m a}{\lambda_p \lambda_S} \frac{\mathcal{G}}{g_b c}. \tag{6}$$

Equation (6) demonstrates a cubic dependence of the Raman gain threshold on the cavity radius a ($V_m \sim a^2$). Experimental studies [21, 22], however, show $\sim a^4$ dependence.

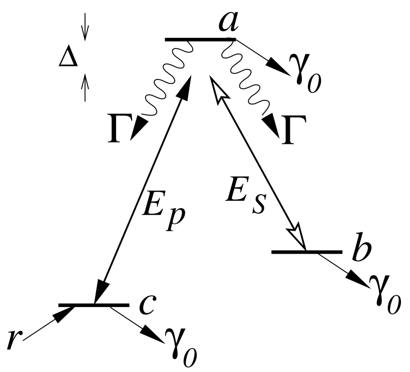

Figure 1. A three-level Λ system coupled by two fields.

To avoid this inconsistency, another Raman gain enhancement factor, proportional to the Purcell factor [1], was introduced heuristically [21] and confirmed theoretically [24]

$$\frac{g_c}{g_b} \approx \frac{3}{4\pi}\frac{\lambda_p^2 c}{V_m \mathcal{G}}. \tag{7}$$

This factor is $\sim\lambda/a$ times as much as the one-dimensional factor (5). The threshold Raman power is here

$$P_{03} > \frac{4\pi^3 n_0^2}{3\xi Q_S Q_p}\frac{V_m^2}{\lambda_p^3\lambda_S}\frac{\mathcal{G}}{g_b c}. \tag{8}$$

In what follows we show, with the example of a three-level Λ Raman medium, that equation (4), along with condition $g_c = g_b$, not equations (5)–(8), does describe the SRS threshold in a microcavity, and propose a classical hypothetical explanation of experimentally observed [21, 22] SRS threshold behaviour.

3. A model of the active Raman system

To compare the Raman gain for unconfined SRS and SRS in a high-Q microcavity, we consider a three-level atomic system in Λ configuration acting as a Raman medium (see figure 1). The electro-dipole allowed transitions of the atom, $|a\rangle \to |c\rangle$ and $|a\rangle \to |b\rangle$, are driven by a strong pump field E_p and a weak probe (Stokes) field E_S, respectively. E_p and E_S are detuned from the corresponding atomic transitions on values $\Delta + \delta$ and $\Delta - \delta$, respectively. In what follows we assume that $|\Delta| \gg |\delta|$. The population of the excited atomic level $|a\rangle$ decays in a free space to the levels $|b\rangle$ and $|c\rangle$ with rate Γ ($|\Delta| \gg \Gamma$).

We consider an open atomic system. The Raman population inversion in the medium is achieved via pumping of the atoms in state $|c\rangle$ into the interaction region and subsequent, time delayed, removal of the atoms from the region. The pumping and removing may be formally introduced by rates r and γ_0. Ratio r/γ_0 determines an average number $\mathcal{N}$ of atoms participating in the interaction.

A coherent evolution of the atom may be described in slowly-varying amplitude and phase approximations by Hamiltonian

$$\hat{H} = \hbar\Delta|a\rangle\langle a| + \hbar\delta(|b\rangle\langle b| - |c\rangle\langle c|) - \hbar(|a\rangle\langle b|\Omega_S + |a\rangle\langle c|\Omega_p + \text{adj}), \tag{9}$$

where $\Omega_S = \wp\hat{E}_S/\hbar$ and $\Omega_p = \wp\hat{E}_p/\hbar$ are the Rabi frequencies of the Stokes and pump fields, $\wp$ is the dipole moment of the allowed atomic transitions and adj denotes the adjoint. In the case of cavity QED interaction, it is convenient to present the amplitudes of the electromagnetic fields via dimensionless creation and annihilation operators. For example, for the pump field, we have

$$\hat{E}_p = \sqrt{\frac{2\pi\hbar\omega_p}{V_p}}\hat{a}_p, \tag{10}$$

where ω_p is the carrier frequency of the pump field, V_p is an effective mode volume for the pump field, and $\hat{a}_p$ is the annihilation operator. Note that we are considering a circular wave in the cavity, not a standing wave. This gives $\sqrt{2}$ difference in κ compared to the conventional standing wave case [28].

It is convenient to introduce the parameter

$$\kappa = \sqrt{\frac{2\pi\wp^2\omega_p}{V_p\hbar}} \tag{11}$$

which describes a strength of coupling of the atom and a cavity and is essential for an explanation of modification of atomic spontaneous emission in the cavity. We assume that Γ is the radiative decay rate so that $\Gamma = 4\omega_p^3\wp^2/(3c^3\hbar)$. In what follows we assume that the cavity modes are nearly identical, so that $\omega_p \simeq \omega_S = \omega_0$, $V_p \simeq V_S = V_m$.

4. Raman amplification in transient regime

To study Raman amplification in a free space we first find a steady state solution for the set of equations for elements of atomic density matrix ρ_{ij}. The coherent part of the set is generated by Hamiltonian (9) as $\mathrm{i}\hbar\dot{\rho} = [\hat{H}, \rho]$, where $\rho = \sum \rho_{ij}|i\rangle\langle j|$. The decays are introduced via general reservoir theory in the Weisskopf–Wigner approximation [28]. We substitute the matrix element of atomic polarization ρ_{ab} into Maxwell equations for the Stokes field and find the Raman gain.

The propagation equation in slowly-varying amplitude and phase approximations may be presented as

$$\frac{\mathrm{d}}{\mathrm{d}z}E_S(z) = \mathrm{i}\frac{2\pi\omega_0}{cV}\mathcal{N}\wp\rho_{ab}, \tag{12}$$

where $\mathcal{N} = r/\gamma_0$ is the total number of atoms in the interaction region and V is the volume of the interaction region.

The evolution of the Λ-system is described by the set of density matrix equations, which includes equations for polarizations

$$\dot{\rho}_{ab} + (\mathrm{i}\Delta + \Gamma + \gamma_0)\rho_{ab} = \mathrm{i}\Omega_S(\rho_{bb} - \rho_{aa}) + \mathrm{i}\Omega_p\rho_{cb}, \tag{13}$$

$$\dot{\rho}_{ca} + (-\mathrm{i}\Delta + \Gamma + \gamma_0)\rho_{ca} = \mathrm{i}\Omega_p^*(\rho_{aa} - \rho_{cc}) - \mathrm{i}\Omega_S^*\rho_{cb}, \tag{14}$$

$$\dot{\rho}_{cb} + \gamma_0\rho_{cb} = -\mathrm{i}\Omega_S\rho_{ca} + \mathrm{i}\Omega_p^*\rho_{ab}, \tag{15}$$

and for populations of the system

$$\dot{\rho}_{bb} = -\gamma_0\rho_{bb} + \Gamma\rho_{aa} - \mathrm{i}(\Omega_S\rho_{ba} - \Omega_S^*\rho_{ab}), \tag{16}$$

$$\dot{\rho}_{cc} = \gamma_0(1-\rho_{cc}) + \Gamma\rho_{aa} - \mathrm{i}(\Omega_p\rho_{ca} - \Omega_p^*\rho_{ac}), \quad (17)$$

$$\dot{\rho}_{aa} = -(2\Gamma+\gamma_0)\rho_{aa} + \mathrm{i}(\Omega_S\rho_{ba} - \Omega_S^*\rho_{ab}) + \mathrm{i}(\Omega_p\rho_{ca} - \Omega_p^*\rho_{ac}), \quad (18)$$

where we have assumed $\delta = 0$. The normalization condition for the population of atomic levels is

$$\rho_{aa} + \rho_{bb} + \rho_{cc} = 1. \quad (19)$$

The solution of equations (13)–(19) completely describes the behaviour of the Λ-system.

The steady state solution of equations (13)–(15) for atomic polarization ρ_{ab} is

$$\rho_{ab} = \frac{\mathrm{i}\Omega_S}{\tilde{\Gamma}} \frac{\left(\gamma_0 + \frac{|\Omega_S|^2}{\tilde{\Gamma}^*}\right)n_{ba} - \frac{|\Omega_p|^2}{\tilde{\Gamma}^*}n_{ca}}{\gamma_0 + \frac{|\Omega_S|^2}{\tilde{\Gamma}^*} + \frac{|\Omega_p|^2}{\tilde{\Gamma}}}, \quad (20)$$

where $\tilde{\Gamma} = \mathrm{i}\Delta + \Gamma + \gamma_0$ and $n_{ij} = \rho_{ii} - \rho_{jj}$. The equation for the atomic polarization on the pump transition may be derived in a similar way.

We are interested here in unsaturated Raman amplification of the Stokes wave, so we assume that the detuning Δ is large, such that $\gamma_0|\Delta| \gg |\Omega_p|^2 \gg |\Omega_S|^2$. Using the above approximations we derive $\rho_{cc} \approx 1$, while $\rho_{aa} \approx 0$ and $\rho_{bb} \approx 0$. The atomic polarization on the Stokes transition may be estimated as

$$\rho_{ab} \simeq -\frac{\mathrm{i}\Omega_S}{\gamma_0}\frac{|\Omega_p|^2}{|\Delta|^2}. \quad (21)$$

Substituting (21) into (12) we arrive at

$$\frac{\mathrm{d}}{\mathrm{d}z}E_S = \frac{3}{8\pi}\frac{r}{\gamma_0}\frac{\lambda_S^2}{V}\frac{\Gamma|\Omega_p|^2}{\gamma_0|\Delta|^2}E_S. \quad (22)$$

Therefore, the intensity of the Stokes wave increases as $I_S(z) = I_S(0)\exp(g_b I_p z)$, where I_p is the intensity of the pump which is connected with the pump Rabi frequency as

$$|\Omega_p|^2 = \frac{3\lambda_p^3\Gamma}{8\pi^2\hbar c}I_p. \quad (23)$$

The bulk Raman gain in the case of the far detuned Λ system is given by

$$g_b = \left(\frac{3}{4\pi}\right)^2 \frac{r}{\gamma_0}\frac{\lambda_S^2\lambda_p^2}{V}\frac{\Gamma^2}{|\Delta|^2}\frac{1}{\hbar\omega_S\gamma_0}. \quad (24)$$

Let us estimate now the power of the Stokes field generated in the SRS process. We consider the one-dimensional case, for example, SRS in a fiber that contains an active Raman medium. Equations for the pump and Stokes modes propagating in the fiber may be presented in the following way [29]

$$\frac{\mathrm{d}}{\mathrm{d}z}P_p(z) = -\alpha_p P_p(z) - \sum_{j=1}^{N_S}\frac{g_b}{\mathcal{A}}\frac{\omega_p}{\omega_{Sj}}P_p(z)P_{Sj}(z), \quad (25)$$

$$\frac{\mathrm{d}}{\mathrm{d}z}P_{Sj}(z) = -\alpha_S P_{Sj}(z) + \frac{g_b}{\mathcal{A}}P_p(z)P_{Sj}(z), \quad (26)$$

where $P_p(z)$ and $P_{Sj}(z)$ are the values of the power of the pump wave and jth mode of Stokes wave, N_S is the total number of Stokes modes excited in the process, α_p and α_S are decays for the pump and the Stokes waves, ω_p and ω_{Sj} are pump and Stokes frequencies, and g_b is the bulk Raman gain coefficient of the medium.

To find the threshold pump power necessary for substantial Raman scattering we assume that there is no nonlinear depletion of the pump so

$$P_p(z) \simeq P_p(0)\mathrm{e}^{-\alpha_p z}. \quad (27)$$

Then

$$P_{Sj}(z) = P_{Sj}(0)\exp\left\{-z\alpha_S + \frac{g_b P_p(0)}{\mathcal{A}\alpha_p}[1-\exp(-z\alpha_p)]\right\}. \quad (28)$$

Assuming that the length of the fiber exceeds the effective absorption length of the pump $L_{eff} = \alpha_p^{-1}$ and $\alpha_p \approx \alpha_S$ we finally get

$$P_{Sj}(L) \simeq P_{Sj}(0)\exp\left[\frac{g_b P_p(0)}{\mathcal{A}\alpha_p} - 1\right]. \quad (29)$$

The Stokes field starts to grow if

$$\frac{g_b P_p(0)}{\mathcal{A}\alpha_p} > 1. \quad (30)$$

This is a conventional definition of the SRS threshold in a transient regime.

However, the Stokes field generated from electromagnetic vacuum fluctuations may be negligible if the pump power is small enough. The critical pump power $P_p(0)$ when the SRS process becomes important may be evaluated from

$$\sum_{j=1}^{N_S}P_{Sj}(0)\exp\left[\frac{g_b P_p(0)}{\mathcal{A}\alpha_p}\right] = \zeta P_p(0), \quad (31)$$

where ζ is a conversion ratio $P_S(L_b)/P_p(L_b)$.

The initial Stokes power may be estimated using an assumption that any Stokes mode, longitudinal as well as transverse, has an input flux of one photon per mode [29]. So,

$$\sum_{j=1}^{N_S}P_{Sj}(0) \approx \hbar\omega_S\frac{\mathcal{A}n_0^2}{\pi^2\lambda_s^2}\left[\frac{\pi\mathcal{A}\alpha_p}{4g_b P_p(0)}\right]^{1/2}\gamma_g, \quad (32)$$

where γ_g is the full width at half maximum of a Lorentzian fit of the Raman gain profile. For the open Λ configuration considered above, $\gamma_g = \gamma_0$. For example, for a single mode fiber, equation (31) transforms to

$$\zeta\left[\frac{g_b P_p(0)}{\mathcal{A}\alpha_p}\right]^{3/2}\exp\left[-\frac{g_b P_p(0)}{\mathcal{A}\alpha_p}\right] = \hbar\omega_S\frac{\sqrt{\pi}}{2}\frac{g_b\gamma_g}{\mathcal{A}\alpha_p}. \quad (33)$$

Generally, condition (30) is assumed to be a threshold for Raman generation in a transient regime. We expect that this is only the necessary condition for the generation. The threshold value for the pump power is to be found from (33). For example, for SRS in silica fiber and for $\zeta = 1$, the pump power is much larger than the threshold when the pump exceeds absorption: $g_b P_p(0)/(\mathcal{A}\alpha_p) \geqslant 20$ [29].

Let us find now the Raman gain in the cavity QED case.

5. Raman amplification in a cavity

The interaction of the cavity electromagnetic fields and the atoms in the cavity may be described in a similar way to the above. However, we should consider now the total number of atoms interacting with the field. Atoms are independent in the case of interaction in free space and, as a result, the electromagnetic susceptibility of the atomic medium is a linear function of the number of atoms participating in the interaction. We find the solution of the interaction problem with the help of the usual density matrix equations (13)–(19). On the other hand, atoms in a cavity influence each on the other, and the method of solution should be modified appropriately. The susceptibility may become a nonlinear function of the number of atoms in the case of cavity-mediated interaction [30].

It is convenient to rewrite the Hamiltonian for a Λ atom in a different form [31, 32]:

$$\tilde{H} = \hbar\left[2\delta - \frac{\kappa^2}{\Delta}(\hat{a}_S^\dagger \hat{a}_S - \hat{a}_p^\dagger \hat{a}_p)\right]|b\rangle\langle b| + \hbar\frac{\kappa^2}{\Delta}(\hat{a}_S^\dagger \hat{a}_p|b\rangle\langle c| + \hat{a}_p^\dagger \hat{a}_S|c\rangle\langle b|), \tag{34}$$

where we used equations (10) and (11) and assumed that Δ is large. The excited state $|a\rangle$ was adiabatically eliminated [33].

To proceed further we derive a set of semiclassical rate equations

$$\dot{n}_S = -2\gamma_S n_S + \tilde{\kappa}\mathcal{N}[n_p(n_S+1)\sigma_{cc} - n_S(n_p+1)\sigma_{bb}], \tag{35}$$

$$\dot{n}_p = -2\gamma_p n_p - \tilde{\kappa}\mathcal{N}[n_p(n_S+1)\sigma_{cc} - n_S(n_p+1)\sigma_{bb}] + \frac{4P_p}{\hbar\omega_p}, \tag{36}$$

$$\dot{\sigma}_{cc} = \gamma_0(1-\sigma_{cc}) - \tilde{\kappa}[n_p(n_S+1)\sigma_{cc} - n_S(n_p+1)\sigma_{bb}], \tag{37}$$

$$\dot{\sigma}_{bb} = -\gamma_0\sigma_{bb} + \tilde{\kappa}[n_p(n_S+1)\sigma_{cc} - n_S(n_p+1)\sigma_{bb}], \tag{38}$$

where

$$\tilde{\kappa} = \frac{2\kappa^4}{\Delta^2\gamma_0}, \tag{39}$$

n_p and n_S are the averaged photon numbers in the pump and Stokes modes, σ_{cc} and σ_{bb} are the averaged collective populations of the corresponding atomic levels and $2\gamma_S = c\alpha_S$. Equations (35)–(38) are to be compared with the rate equations for a four-level microlaser [3, 34–36]. The quantum modification of (35)–(38) might be easily seen as the unity factors added to the photon numbers n_S and n_p. This change modifies the behaviour of the system close to threshold and/or when the system operates with very weak pump. The Raman microlaser is thresholdless in the same sense as the usual microlasers [3, 34–36]. For example, the number of Stokes photons is never zero if $\sigma_{cc} \approx 1$ and $P_p \neq 0$.

In the case where the drive field is not influenced by the Stokes field ($n_p \gg n_S$) and $\sigma_{cc} \approx 1$ the threshold condition for the exponential growth of the Stokes field formally coincides with (30) and condition (4):

$$\frac{\tilde{\kappa}\mathcal{N}}{2\gamma_S}n_p = \frac{g_b P_{pc}}{\mathcal{A}\alpha_p} > 1, \tag{40}$$

where we assumed that $\alpha_p = \alpha_S$ and P_{pc} is the effective pump power inside the cavity, $P_{pc}/P_p = Q\lambda/(\pi^2 a)$. This is the main result of the paper. There is no cavity QED increase of the Raman gain. Hence, condition (4) is correct for the description of the SRS in a cavity.

It is interesting now to look at the threshold condition for Raman lasing in the sense of equation (33), because, generally, SRS is observed when the numbers of generated and pump photons are comparable. We derive two steady state balance equations from (35)–(38):

$$n_p = \frac{4P_pQ_p}{\hbar\omega_p^2} - \frac{\gamma_S}{\gamma_p}n_S, \tag{41}$$

$$\sigma_{cc} = 1 - \frac{2\gamma_S}{\mathcal{N}\gamma_0}n_S, \tag{42}$$

where (41) shows that the total number of photons leaving the cavity is equal to the number of photons entering the cavity, and (42) shows that number of signal photons leaving the cavity ($2\gamma_S n_S$) is always less than the number of atoms entering the cavity ($\gamma_0\mathcal{N} = r$).

Let us assume that $\gamma_S = \gamma_p$ and $n_S = n_p \gg 1$ ($\zeta = 1$). The Stokes photon number and atomic pumping rate r may be found from the steady state solution of (35) along with (41) and (42):

$$n_S = \left(1 - \frac{2\mathcal{A}\alpha_p}{g_bP_{pc}}\right)\frac{r}{2\gamma_S} = \frac{2P_pQ_p}{\hbar\omega_p^2}. \tag{43}$$

Therefore, because the photon number is positive ($n_S \geqslant 0$), the effective gain factor $g_bP_{pc}/(\mathcal{A}\alpha_p)$ should be more than 2, independent of the pumping rate r. This is an order of magnitude less than the gain value for maintaining $\zeta = 1$ in the transient regime, even after we took into account the power increase in the cavity. This might be the reason why the experimentally observed Raman threshold in a cavity is lower than the threshold observed for the transient regime. The result, however, is rather classical and it does not explain the dependence of the Raman gain on the cavity size measured in [21, 22].

6. Size dependence of SRS threshold: a hypothesis

As shown above, Raman gain is not enhanced due to cavity QED effects ($g_b = g_c$). The question is, how to explain the cavity size dependent Raman gain behaviour (SRS threshold decreases as $\sim a^4$) observed in [21, 22]. To begin with, we modify the threshold condition (4) as

$$P_0 > \frac{\pi^2 n_0^2 Q_{Pc}}{\xi_1 g_b Q_S Q_p^2}\frac{V_m}{\lambda_p\lambda_S}, \tag{44}$$

where $\xi_1 < 1$ is a parameter indicating that mode overlap is not complete, and Q_{Pc} is a part of cavity quality factor that results from the coupling ($Q_{Pc} > Q_p$). For the critical coupling one has $Q_{Pc} = 2Q_p$. Cavity parameters ξ_1, V_m, Q_p, Q_S, and Q_{Pc} are size dependent, while g_b is not.

Let us estimate the changes of these parameters for an interaction of a dielectric sphere with a free beam of light. First and foremost, there is no critical coupling with a high order WGM in this case. Ratio Q_{Pc}/Q_p is always much larger than 2. For example, let us consider radiative coupling with a microsphere. This kind of coupling prevails for small microspheres, where quality factor is determined by radiative decay rate.

Radiative coupling of a microsphere and a beam of light may be described using generalized Lorentz–Mie scattering theory [37–40]. Exact calculation of ratio Q_{Pc}/Q_p is out of the scope of the present paper. We use the modified ray theory described in [41] and confirmed in [42].

Let us consider a light beam interacting with a sphere. The beam cross-section radius is comparable with the sphere radius a (total cross-section $\sim 2\pi a^2$). The scattering cross-sectional mode area is $\sim a\lambda$ [41]. Therefore, only $\sim \lambda/(2\pi a)$ of the total power interacts with the sphere. To estimate Q_{Pc}/Q_p we assume that the radiative losses and optical pumping have the same origin and are proportional to the interaction surface area. The radiative emission occurs from all the sphere surface (area equal to $4\pi a^2$). The optical pumping is going through a surface belt with thickness $\sim\lambda$. Therefore, the ratio of the coupling and total quality factors is proportional to the sphere radius $Q_{Pc}/Q_p \sim a$.

The mode volume for a high order WGM is proportional to $\sim a^2$. However, because a plane wave excites all modes under the microsphere surface, not just a single mode 'belt', the excited volume depends on the higher power of the radius $\sim a^{2+1/3}$. Moreover, because the increase of the microsphere size reduces coupling to the high order WGM, the light may primarily excite the lower order modes for bigger cavities. This may result in even faster growth of the effective mode volume with radius a. Hence, the SRS threshold will decrease faster than $a^{3+1/3}$ if we assume that the system behaviour is determined by radiative processes and the quality factor for each cavity size is nearly the same.

The last assumption, based on the experimental observations, that the measured quality factors for small droplets does not vary significantly with their sizes supports the hypothesis that a free light beam excites different orders of cavity modes with cavity radius increase. Theoretically, the radiative quality factor of an ideal WGM should increase exponentially with the mode radius. Had it been possible to excite these modes all the time, the threshold power (44) would have exponentially decreased (not increased, as in the experiments) with cavity radius decrease.

Generally, the quality factor of a dielectric microcavity is determined not by the radiative tunnelling emission, but by the surface and volume scattering and absorption [43–48]. The surface scattering may aid the coupling of the free light beam and modes. The above estimations and conclusions are valid in this case as well.

All the above inconsistencies and problems with determining which mode is excited, what is the value of the mode volume, and what is the mode quality factor cease if one uses special coupling techniques for the light and the microspheres (see, for example, [25, 44]). As the result, the carefully measured SRS threshold shows square dependence on the cavity radius, which coincides with our theoretical predictions.

7. Conclusion

We have shown theoretically that the Raman gain in an SRS process is associated with the intrinsic properties of the active medium only. This gain cannot be changed via redistribution of the mode density in a cavity. The enhancement of efficiency of the scattering results from energy accumulation in the cavity modes. In general, this conclusion is valid for any nonlinear process, e.g. four-wave mixing. Spontaneous emission, in turn, may be enhanced or suppressed by a cavity. The size dependence, observed for SRS in microdroplets, may result from the size-dependent coupling efficiency with the droplets as well as from size dependent quality factor of the droplets.

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